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NON-MONOTONOUS DEPENDENCES OF THIN FILM SURFACE ROUGHNESS ON SUBSTRATE TEMPERATURE AND DEPOSITED ATOM FLUX

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Experimentally observed non-monotonous dependences of thin films surface roughness on substrate temperature and flux of deposited atoms are analysed by kinetic rate equation model. The modelling results show good qualitative agreement with the experiment and explain an unusual phenomenon of surface roughness. It is shown that non-monotonous dependence of surface roughness on substrate temperature and deposition flux is determined by the size of islands and diffusivity of atoms on the surface. From present analysis it follows that the formation mechanisms of non-monotonous dependences of the surface roughness on temperature and deposition flux are of different origin. The mechanisms are qualitatively analysed in the present paper.

Keywords: surface roughness, island films deposition, surface diffusion, kinetic modelling

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1. Introduction

The surface roughness is one of the most important parameters of thin films that influence growth properties of the deposited films. Smooth surfaces lead to a more uniform, fine grain structure, while rough surfaces with peaks, valleys, and other defects act as either preferred sites or lead to shadowing of the adatoms and more columnar-type microstructure [1]. The morphology and surface roughness of growing thin film can be influenced by substrate relief (the substrate relief influences the cluster formation on the surface) [2], deposited material lattice mismatch with substrate [3], substrate surface crystallographic direction (the diffusion length of adatoms and diffusion coefficient depend on surface direction) [4], etc. The surface roughness depends on many factors during process of thin film deposition. The fundamental physical processes, such as adsorption, desorption, nucleation, surface diffusion, coalescence of islands, heterogeneous reaction, etc., are being observed in early thin film growth stages. All these processes take place simultaneously and determine the surface morphology of the film. Moreover, these processes proceed differently for different materials and depend on the process conditions. Various surface roughness dependences on different technological parameters, such as substrate temperature, ion energy, flux of arriving atoms, etc., have been investigated experimentally [5–12]. The analysis of different factor influence on the surface roughness has shown that the surface roughness can decrease, increase, or stay constant depending on considered interval of atom energies.

There are many works where non-monotonous surface roughness dependence on the substrate temperature [5–8] and the flux of arriving atoms [11] have been observed. Several examples are presented in Fig. 1 [6, 11]. In many papers published recently the authors present different explanations of the formation of this minimum point. Earlier it has been shown by Monte Carlo calculations [13] that the diffusion ability of adatoms increases with increase of the substrate temperature and more adatoms can be activated to diffuse and fill in the inner voids of the film. Then surface roughness decreases with temperature. However, when the temperature becomes too high the diffusivity of atoms is very high so that some of them can jump onto the top of upper layer. As a result the surface roughness increases with temperature again [13]. Authors of the

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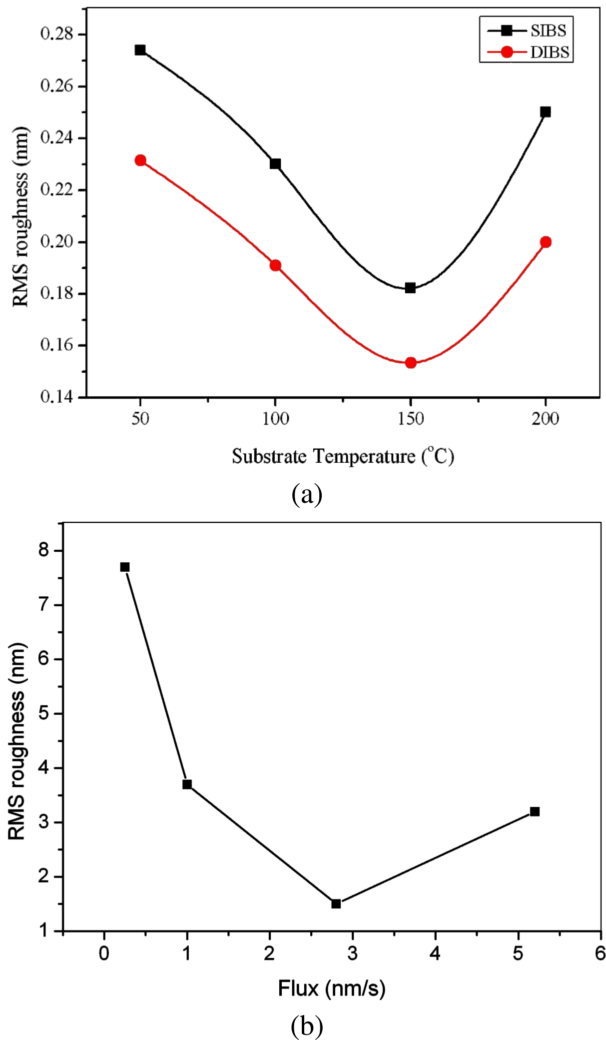

Fig. 1. Dependence of surface roughness on the substrate temperature and on the flux of arriving atoms: (a) experimental data, obtained by Yoon et al. [6] (SIBS: single ion beam sputter deposition, DIBS: dual ion beam sputter deposition), (b) experimental data, obtained by Dumont et al. [11].

Fig. 1(b) [11] explain non-monotonous dependence of the surface roughness on the deposition rate using the relation between the average distance between islands and critical island radius at which a second layer nucleates on the top of these islands (the so-called "TDT approach" theory). The layer-by-layer growth is observed and the surface becomes smoother, if critical island radius is larger than the average distance between the islands. If critical island radius is small compared to the average distance between the islands, they will nucleate a second layer before coalescence, giving multilayer growth. As a result the surface becomes rougher. Despite the many experimental and analytical studies performed, the reason of non-monotonous variation of surface roughness is not clear enough.

The purpose of this work is to explain non-monotonous dependences of the surface roughness as a function of substrate temperature and deposition flux. In our previous work [14] the model was proposed which explains the non-monotonous dependence of surface roughness on diffusivity of adatoms. In this study we consider the influence of substrate temperature and deposition flux to the surface roughness which shows non-monotonous dependences in both cases (observed in many experiments). It is shown in this paper that despite both dependences being mathematically described by the same model, the explanation and physical reasons are different: diffusivity of adatoms strongly depends on substrate temperature (so the explanation is similar to that given previously [14]), but does not depend on deposition flux. To show this difference, both cases are analysed in this paper. For analysis some typical experimental results are taken from literature and qualitatively compared with calculated curves obtained by our model. Theoretical results show a good qualitative agreement with the experimental ones and allow explaining the physical origin of non-monotonous surface roughness variations with the substrate temperature and deposition flux.

2. Kinetic model of island film growth

Film growth can be divided into the following elementary steps: adsorption, diffusion of single atoms on the surface; nucleation of new clusters and coalescence. All these steps can be expressed by kinetic equations defining the time variation of surface coverage φ and cluster density n. The main idea of the model is to separate the coverage of the first monolayer into coverage by single atoms φ_S and by islands φ_C. This gives a possibility to include the processes of island nucleation and growth, coalescence, and other kinetic processes. More detailed description of the model can be found in our previous work [15, 16].

Thus, the kinetics of the first monolayer coverage by the single adatoms φ_S may be obtained as

$$\frac{\mathrm{d}\varphi_S}{\mathrm{d}t} = \alpha_{A0} i_0 (1-\varphi_L) - \alpha_{AA} i_0 \varphi_S \,, \qquad (1)$$

where t is the time, i_0 is the relative flux of arriving single atoms. The surface coverage φ_S increases (the first term) due to the arrival of atom that sticks at the substrate surface with the sticking coefficient α_{A0}. The second term describes the decrease due to cluster nucleation process: an arriving atom sticks at the already existing single adatom with the probability α_{AA} and forms a new cluster of two atoms. $(1-\varphi_L)$ represents un-

covered part of substrate which also involves diffusion length of single atoms λ ($\varphi_L = \varphi_S + \varphi_C + a(t)\varphi_C^*$). φ_C^* is a dimensionless area with radius λ (equal to diffusion length of adatoms) around the clusters within which the arrived atom will stick at the edge of the cluster. Because φ_L involves the term φ_C^* (will be precisely defined later in Eq. (8)) and at higher coverage may exceed 1, the normalization parameter $a(t)$ is introduced, which obtains the following values:

$$a(t) = \begin{cases} 1 & \text{if } \varphi_L < 1\,, \\ \dfrac{1 - \varphi_S(t) - \varphi_C(t)}{\varphi_C^*(t)} & \text{if } \varphi_L \geq 1\,. \end{cases} \quad (2)$$

The equation for the kinetics of substrate coverage by clusters φ_C is the following:

$$\frac{\mathrm{d}\varphi_C}{\mathrm{d}t} = 2\alpha_{A0} i_0 \varphi_S + a(t)\alpha_{AC} i_0 \varphi_C^* + B^{(1)}\alpha_{AC} i_0 (1 - \varphi^{(1)})(\varphi^{(1)} - \varphi^{(2)})\,. \quad (3)$$

The substrate coverage by clusters increases after the nucleation process (the first term), then after the atom sticks at the edge of already existing island with sticking coefficient α_{AC} (the second term), and when the atom jumps down from the top of an island because of the surface diffusion and sticks at the edge of the island (the third term). $B^{(1)}$ is the possibility of atom jumping down from upper monolayer to stick at the edge of island on the first monolayer. $\varphi^{(1)}$, $\varphi^{(2)}$ are the coverages of first and second monolayer, respectively.

The fourth equation presents the kinetics of the coverage of Kth (>1) monolayer $\varphi^{(K)}$:

$$\frac{\mathrm{d}\varphi^{(K)}}{\mathrm{d}t} = A^{(K)}\alpha_{AT} i_0 (\varphi^{(K-1)} - \varphi^{(K)}) + B^{(K)}\alpha_{AT} i_0 (\varphi^{(K)} - \varphi^{(K+1)}) \times (\varphi^{(K-1)} - \varphi^{(K)})\,, \quad K = 2, 3, \ldots\,. \quad (4)$$

The coverage of Kth monolayer increases when the atoms directly arrived remain on it (the first term) or because of the surface diffusion process (the second term) when atoms are jumping down from the upper monolayers. The possibility of the atom to stick on the top of the cluster or to jump down from it is defined by the coefficients $A^{(K)}$ and $B^{(K)}$ [15, 16].

The kinetics of relative islands density n is defined by the equation

$$\frac{\mathrm{d}n}{\mathrm{d}t} = \begin{cases} 0.5\alpha_{AA} i_0 \varphi_S + \alpha_{\text{coal}} C_{\text{coal}} (1 - \varphi_S - \varphi_\beta) & \text{if } r < r_{\text{max}}\,, \\ 0 & \text{if } r \geq r_{\text{max}}\,. \end{cases} \quad (5)$$

The density of islands n increases because of nucleation (the first term) and decreases because of coalescence effect when two islands coalesce into a larger one as they touch each other with the probability α_{coal} (the second term). C_{coal} is normalization factor (exact definition given in Ref. [15]) and $\varphi_\beta = n\beta^2 + 2\beta\sqrt{n\varphi_C}$ is the dimensionless area around the island within the range of the migration radius β of the island. When $\varphi_\beta \neq 0$, the islands can migrate, therefore the second term in Eq. (5) describes mobile coalescence effect.

The islands can contact mutually when the radius of the islands reaches the critical radius r_{max}. It is assumed that at this moment the coalescence process stops (the second condition in Eq. (5)). If this happens the number of clusters remains constant. This also means the process of nucleation has stopped as well. The condition for physical contact of clusters was assumed to be the following:

$$4\,n\,r_{\text{max}}^2 = 1\,. \quad (6)$$

The surface roughness $\delta(t)$ as a function of deposition time is defined as

$$\delta(t) = P_2(t) - P_1(t)\,, \quad (7)$$

where $P_1(t)$ and $P_2(t)$ are the distances expressed in terms of the number of monolayers from the initial surface ($K = 1$) to the monolayer with the coverage equal to $\varphi(P_1(t)) = 0.05$ and $\varphi(P_2(t)) = 0.95$, respectively.

The atoms on the surface, before they are adsorbed, diffuse over the surface. The diffusion length λ is included in the model through the term φ_C^* in Eq. (3), and has the following form:

$$\varphi_C^* = n\lambda^2 + 2\lambda\sqrt{n\varphi_C}\,. \quad (8)$$

Generally, the diffusion length is defined by relation $\lambda = \sqrt{4Dt}$, where D is the diffusion coefficient. The diffusion length $\lambda(T_{\text{sub}})$ as a function of the substrate temperature T_{sub} can be expressed from Arrhenius law for diffusion $D = D_0 \exp[-E_{\text{a}}/(kT_{\text{sub}})]$:

$$\lambda = 2\sqrt{D_0 \exp\left(\frac{-E_{\text{a}}}{kT_{\text{sub}}}\right) t}\,, \quad (9)$$

where D_0 is the preexponential constant, E_{a} is the surface diffusion activation energy, k is the Boltzmann

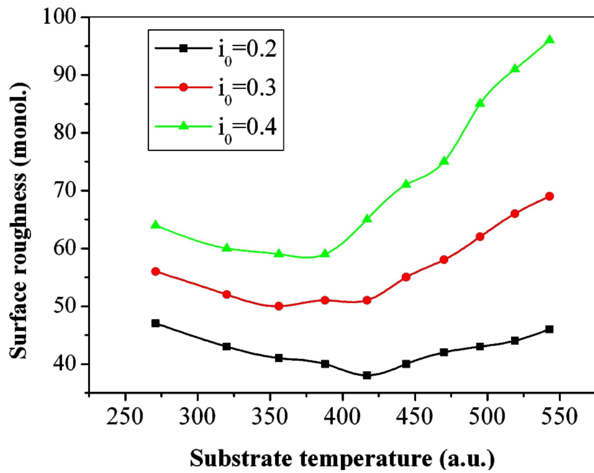

Fig. 2. Dependences of surface roughness on the substrate temperature in the cases of different flux of deposited atoms i_0. Coefficients used in calculations: $\alpha_{A0} = 0.1$, $\alpha_{AA} = 0.0005$, $\alpha_{AC} = 0.1$, $\alpha_{AT} = 1.0$, $\alpha_{\text{coal}} = 0.0001$, $C = 1$, $\beta = 20$, $\lambda = \lambda_T$.

constant. The expression gives the increase of diffusion length with the substrate temperature.

3. Results and discussion

Single atoms arriving to the substrate diffuse over the surface until they nucleate into the new island or are trapped by existing islands. The nucleation of new islands depends on the surface diffusion activation barrier E_a and on the substrate temperature T_sub (Eq. (9)).

The experimental results from Refs. [6, 11] presented in Fig. 1(a) show that the dependence of the surface roughness on the substrate temperature can be non-monotonous. The modelling results presented in Fig. 2 show that the curve of surface roughness dependence on the substrate temperature passes a minimum value point when the temperature is increased. Figure 2 shows that our modelling results are qualitatively in a good agreement with experimental results of Fig. 1(a).

The non-monotonous dependence of the surface roughness on the substrate temperature can be explained by interplay between the island size and the diffusion length of adatoms (Fig. 3). The adatom diffusivity and the island sizes are negligible at low temperatures. Despite the small size of the islands, diffusivity of the adatoms is too low and the adatom cannot jump down from the top of islands. The adatoms arrived to the top of the island stick on it in this case. As a result the surface becomes rougher. The diffusivity of the adatoms and the size of islands increase as the temperature increases, yet the diameter of islands does not increase so fast and the atoms that have climbed onto the top of the islands could jump down to deeper layers. In this case

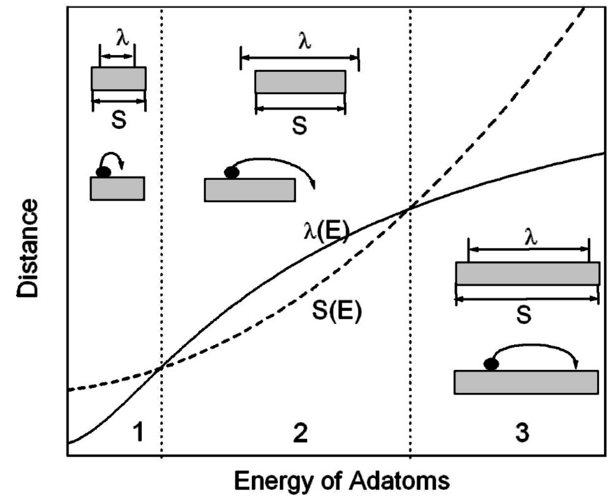

Fig. 3. Schematic drawing of the island size and diffusion length of adatoms and interplay between them, which explains the formation of non-monotonous dependence of surface roughness versus energy of the adatoms.

the surface becomes smoother. The size of the islands may exceed the adatom diffusion length again at higher temperatures. The atoms arrived to the island top layer remain on it and the surface roughness increases again.

The study of the surface roughness of a growing thin film as a function of substrate temperature and flux of arriving atoms was performed using the above kinetic model in two cases:

(i) the diffusion length of atoms on the pure substrate, λ, and on the deposited material, λ_T, are equal ($\lambda = \lambda_T$);

(ii) the diffusion length of atoms on the pure surface, λ, is significantly higher than the diffusivity of adatoms on deposited material, λ_T ($\lambda \gg \lambda_T$).

It is well known that at random deposition (random sticking of atoms) [17] the surface roughness δ increases with deposition time t as its square root, $\delta \propto \sqrt{t}$. The exact expression for the surface roughness can be found in that case, $\delta \propto \sqrt{4\kappa t}$, where κ is the adsorption rate: $\kappa = \alpha\, i_0$ (α is sticking coefficient and i_0 is atomic flux). The intensity of flux acts in a similar way as duration of the deposition time: the number of the adsorbed atoms increases in both cases. Therefore, it follows that the surface roughness δ increases with the flux i_0 in the same manner as with the deposition time, i. e. $\delta \propto \sqrt{i_0}$. The calculated dependences of the surface roughness on the deposition flux in the case of $\lambda = \lambda_T$ are shown in Fig. 4, where the presented curves follow the law $\delta \propto \sqrt{i_0}$, i. e. the surface roughness monotonously increases with the deposition flux. However, the law $\delta \propto \sqrt{i_0}$ is valid for random

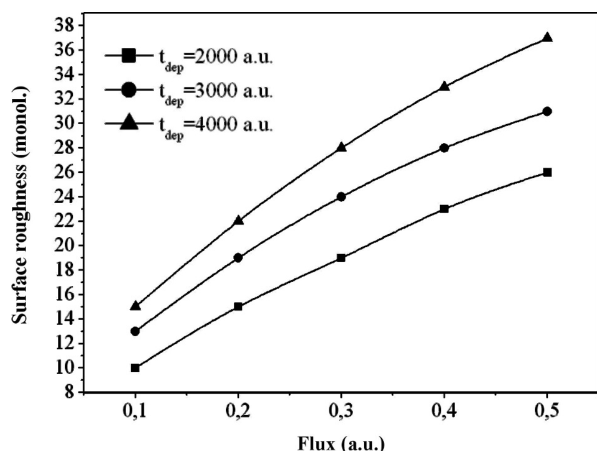

Fig. 4. Dependence of surface roughness on the flux of arriving atoms in the case $\lambda = \lambda_T$. Coefficients used in calculations: $\alpha_{A0} = 0.05$, $\alpha_{AA} = 0.005$, $\alpha_{AC} = 0.1$, $\alpha_{AT} = 1.0$, $\alpha_{\text{coal}} = 0.001$, $C = 1$, $\beta = 15$, $\lambda = \lambda_T = 2$.

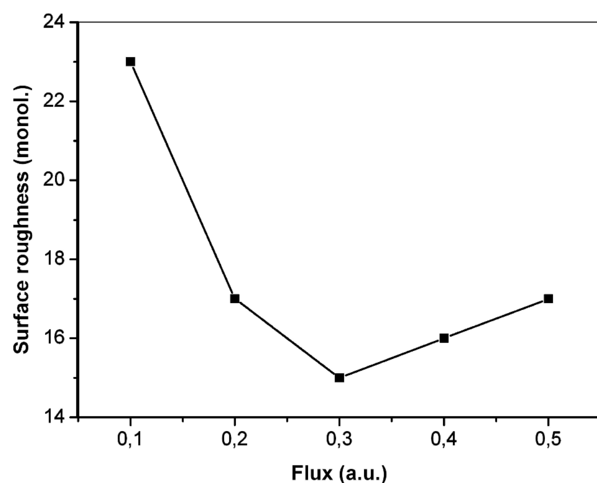

Fig. 5. Dependence of surface roughness on the flux of arriving atoms in the case $\lambda \gg \lambda_T$. Coefficients used in calculations: $\alpha_{A0} = 0.05$, $\alpha_{AA} = 0.001$, $\alpha_{AC} = 0.1$, $\alpha_{AT} = 1.0$, $\alpha_{\text{coal}} = 0.0001$, $C = 1$, $\beta = 25$, $\lambda = 30$, $\lambda_T = 1$.

(no islands) sticking of arriving atoms only. In the case of driven sticking the deviation from this law is possible. The driven sticking means that the adsorption of atoms may be influenced by some driving forces under which some geometrical structures are formed. Island film growth belongs to the latter type.

The calculated dependence of the surface roughness on the deposition flux in the case of $\lambda \gg \lambda_T$ is presented in Fig. 5. The character of the curve is completely different from that presented in Fig. 4. The dependence is non-monotonous: the roughness decreases at the low fluxes and increases at the higher fluxes. The results of Fig. 5 are confirmed by Dumont's et al. experimental observations [11] for thin silver films deposited on mica substrate (Fig. 1(b)). The experimentally mea-

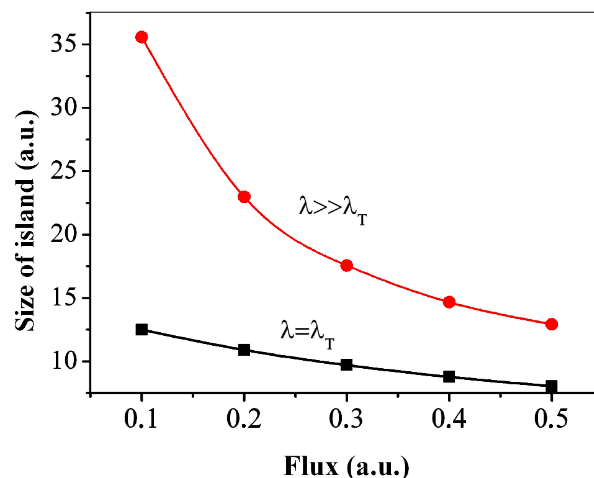

Fig. 6. Dependences of island size on the deposition flux in two cases, $\lambda = \lambda_T$ and $\lambda \gg \lambda_T$.

sured dependence of surface roughness on deposition rate shows the same behaviour (Fig. 1(b)).

As follows from the presented model, the decrease of roughness with increase of deposition flux occurs because of atomic jumps from the top of islands when diffusion length exceeds the size of island (regime *2* in Fig. 3). As shown in Fig. 6, where calculated dependences of the island size on deposition flux are presented, the size of islands decreases with increase of the deposition flux. The both curves, $\lambda = \lambda_T$ and $\lambda \gg \lambda_T$, show the same behaviour, but the island size in the case $\lambda \gg \lambda_T$ is significantly higher. In the case of larger islands, the decrease of island size gives more intense jumps of atoms from the top of islands to the substrate. As a result, the surface roughness decreases. However, the random sticking defined by the law $\delta \propto \sqrt{i_0}$ prevails again when islands become too small. The surface roughness starts to increase as it is seen in Fig. 5. Random sticking always takes place, tending to increase the roughness, whereas driven sticking is just an additional term which in the case of large islands reduces this tendency. Then the deviation from general law $\delta \propto \sqrt{i_0}$ occurs.

4. Conclusions

1. From the presented analysis the explanation of non-monotonous dependence of surface roughness on temperature is the following: (i) in the low temperature region the surface roughness decreases with temperature because the small island formation and atomic jumps from the top of islands onto the substrate prevail in this case; (ii) in the high temperature region large islands are formed and arriving atoms stick on the top of

the islands, causing the surface roughness to increase with temperature.

2. The explanation of non-monotonous dependence of surface roughness on the deposition flux is the following. There is a general rule that the surface roughness at random deposition (no islands) increases with the deposition flux. However, there is a deviation from this rule if large islands are formed. Island size decreases with increasing deposition flux and the jumps of atoms down from the top of an island to the substrate become more intense. Then the decrease of the surface roughness takes place. At high flux, very small islands are formed and the jumps of atoms from the top of islands are not important. The random deposition regime prevails and surface roughness starts to increase with deposition flux.

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NEMONOTONINĖS PLONŲ DANGŲ PAVIRŠIAUS ŠIURKŠTUMO PRIKLAUSOMYBĖS NUO PADĖKLO TEMPERATŪROS IR NUSODINTŲ ATOMŲ SRAUTO

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Santrauka

Paviršiaus šiurkštumas yra vienas iš svarbiausių parametrų, apibūdinančių plonas dangas. Yra daug eksperimentinių darbų, kuriuose nagrinėjama plonų dangų paviršiaus šiurkštumo priklausomybė nuo įvairių technologinių dangos auginimo parametrų: padėklo temperatūros, jonų energijos, dalelių srauto ir kt. Apibendrinant šiuos darbus, galima pasakyti, kad plonų dangų paviršiaus šiurkštumas, priklausomai nuo technologinių parametrų kitimo, gali didėti, mažėti ar nekisti. Taip pat yra darbų, kuriuose stebima nemonotoninė paviršiaus šiurkštumo priklausomybė nuo padėklo temperatūros [6] bei dalelių srauto (nusodinimo greičio) [11]: paviršiaus šiurkštumo priklausomybė nuo šių dydžių turi minimumo tašką.

Pasinaudojant kinetiniu modeliu [15, 16], tirta plonų dangų paviršiaus šiurkštumo priklausomybė nuo padėklo temperatūros bei į paviršių krintančių dalelių srauto. Panaudotas kinetinis modelis, aprašantis salelinį dangos augimą ir apimantis tokius procesus kaip adsorbuotų atomų paviršinė difuzija, nukleacija, susidariusių salelių augimas bei koalescencija. Modeliavimo rezultatai parodė, kad ir padėklo temperatūra, ir dalelių srautas veikia ant padėklo susidarančių salelių dydį, tuo pačiu ir plonų dangų paviršiaus šiurkštumą. Gauti modeliavimo rezultatai gerai sutampa su eksperimentiniais ir leidžia paaiškinti nemonotoninį paviršiaus šiurkštumo kitimą.